

**NON-THERMAL PLASMA APPARATUS UTILIZING
DIELECTRICALLY-COATED ELECTRODES FOR TREATING EFFLUENT GAS**

5

CONTRACTUAL ORIGIN OF THE INVENTION

The United States Government may have rights in this invention pursuant to the Cooperative Research And Development Agreement ("CRDA") between Science Applications International Corporation and the Air Force Wright Laboratory Armament Directorate.

10

FIELD OF THE INVENTION

This invention relates to an improved apparatus for treatment of hazardous gases, in particular, an improved non-thermal plasma apparatus for the removal of NO_x from gas streams resulting from various chemical processes and by the combustion of carbonaceous fuels.

20

BACKGROUND AND SUMMARY OF THE INVENTION

The current global concern for the environment has generated a strong need by both government and industry for technologies that reduce emissions of NO_x. NO_x are primary contributors to photochemical smog and acid rain, and may deplete the ozone layer. Up to millions of tons of nitrogen oxides, generally denoted herein as "NO_x," are emitted into the atmosphere each year as a result of numerous industrial and

military processes, ranging from high temperature combustion of fossil fuels, to explosive manufacturing and munitions disposal processing and further to operations of powered aerospace ground equipment.

5

The impact of such emissions on human health and the environment in general has been the subject of intense study and public debate and legislative action to mandate safer emissions has already been enacted. For example, the Clean Air Act Amendment of 1990 mandates that emission generating industrial plants develop and/or implement techniques to significantly reduce their emissions of NO_x. Such legislation affects power plants, iron and steel plants, pulp and paper mills, acid production plants, petroleum refineries, lime plants, fuel conversion plants, glass fiber processing plants, charcoal production plants, cement plants, copper smelters, coal cleaning plants, etc.

20 converting NO_x to the individual elemental diatoms, N₂ and O₂. Conventional processes typically utilized thermal techniques for generating very high temperature conditions within a reactor. These techniques are highly inefficient as excessively high electrical power is needed not only to treat 25 relatively low pollutant concentrations, but to cool the resultant effluent emerging from the reactor.

Electron beam irradiation has also been used in various forms to convert to the individual elemental diatoms. Such systems also use electron beams or ultraviolet light to oxidize the NO_x. The ionization caused by the electron beam irradiation converts the NO_x to acid mist at low temperatures and/or solid particles at high temperatures which may be removed by conventional methods employing filters and scrubbers. However, due to potential harm to operation personnel, costly and elaborate shielding measures must be employed.

Numerous research agencies have investigated the use of non-thermal plasma devices ("NTPDs") to reduce NO_x in gas streams. These devices employ electrodes and dielectrics driven by a voltage supply for generating an electric field driving electro-chemical reactions converting NO_x to other atomic and molecular forms, either chemically less toxic, and/or structurally more readily removed from the gas stream. Numerous materials have been used to construct such dielectrics, including quartz, glass, alumina, mullite, and oxide free ceramic such as silicon nitrite, boron nitrite, aluminum nitrite.

Of the conventional non-thermal plasma devices, the dielectrics isolating the electrodes from the gas and enabling the non-thermal plasma environment which drives various

electro-chemical reactions are typically constructed of glass. While glass has the required thermal and electrical properties, it fractures easily and may not be well suited for any mobile applications of non-thermal plasma devices. In addition, an appreciable thickness of glass is typically used to standoff the voltages applied, which limits the flow rate or throughput of the devices. As for the other conventional dielectric materials, many have suitable dielectric strength and many have suitable working temperatures; however, none combine both of these factors in providing a desirable dielectric for use in an efficient effluent gas treating non-thermal plasma apparatus.

Accordingly, there exists a demand for an improved non-thermal plasma apparatus for treating NO_x bearing gas streams, with significantly improved efficiency and durability. It is also desired that such an apparatus has significantly increased applicability and ease of application. In accordance with the present invention, a non-thermal plasma apparatus is provided, having a plasma reactor and an inlet and outlet connected thereto through which the exhaust gas enters and leaves the plasma reactor. A scrubber may be provided to scrub the exhaust gas leaving the plasma reactor and a stack may be connected to the scrubber to permit the exhaust gas to exit the apparatus. The plasma reactor is equipped with a plurality of dielectrically-coated electrodes between which a selected voltage is applied to generate a non-thermal plasma environment.

5

for driving selected electro-chemical reactions. A voltage supply is electrically configured to apply a predefined voltage across the electrodes to create microdischarges in the exhaust gas stream. Where such electro-chemical reactions involve the conversion of nitric oxides to various diatomic and molecular forms, including primarily nitrogen dioxide, the efficiency (in terms of energy per molecule of remediated NO_x) and total reduction (in terms of percentage of hazardous compounds reduced per treatment pass) may be increased by approximately 30% when compared with conventional non-thermal plasma devices.

202
150
152
154
155
156
157
158
159
160
161
162
163
164
165
166
167
168
169
170
171
172
173
174
175
176
177
178
179
180
181
182
183
184
185
186
187
188
189
190
191
192
193
194
195
196
197
198
199
200
201
202
203
204
205
206
207
208
209
210
211
212
213
214
215
216
217
218
219
220
221
222
223
224
225
226
227
228
229
230
231
232
233
234
235
236
237
238
239
240
241
242
243
244
245
246
247
248
249
250
251
252
253
254
255
256
257
258
259
260
261
262
263
264
265
266
267
268
269
270
271
272
273
274
275
276
277
278
279
280
281
282
283
284
285
286
287
288
289
290
291
292
293
294
295
296
297
298
299
300
301
302
303
304
305
306
307
308
309
310
311
312
313
314
315
316
317
318
319
320
321
322
323
324
325
326
327
328
329
330
331
332
333
334
335
336
337
338
339
340
341
342
343
344
345
346
347
348
349
350
351
352
353
354
355
356
357
358
359
360
361
362
363
364
365
366
367
368
369
370
371
372
373
374
375
376
377
378
379
380
381
382
383
384
385
386
387
388
389
390
391
392
393
394
395
396
397
398
399
400
401
402
403
404
405
406
407
408
409
410
411
412
413
414
415
416
417
418
419
420
421
422
423
424
425
426
427
428
429
430
431
432
433
434
435
436
437
438
439
440
441
442
443
444
445
446
447
448
449
4410
4411
4412
4413
4414
4415
4416
4417
4418
4419
4420
4421
4422
4423
4424
4425
4426
4427
4428
4429
4430
4431
4432
4433
4434
4435
4436
4437
4438
4439
4440
4441
4442
4443
4444
4445
4446
4447
4448
4449
44410
44411
44412
44413
44414
44415
44416
44417
44418
44419
44420
44421
44422
44423
44424
44425
44426
44427
44428
44429
44430
44431
44432
44433
44434
44435
44436
44437
44438
44439
44440
44441
44442
44443
44444
44445
44446
44447
44448
44449
444410
444411
444412
444413
444414
444415
444416
444417
444418
444419
444420
444421
444422
444423
444424
444425
444426
444427
444428
444429
444430
444431
444432
444433
444434
444435
444436
444437
444438
444439
444440
444441
444442
444443
444444
444445
444446
444447
444448
444449
4444410
4444411
4444412
4444413
4444414
4444415
4444416
4444417
4444418
4444419
4444420
4444421
4444422
4444423
4444424
4444425
4444426
4444427
4444428
4444429
4444430
4444431
4444432
4444433
4444434
4444435
4444436
4444437
4444438
4444439
4444440
4444441
4444442
4444443
4444444
4444445
4444446
4444447
4444448
4444449
44444410
44444411
44444412
44444413
44444414
44444415
44444416
44444417
44444418
44444419
44444420
44444421
44444422
44444423
44444424
44444425
44444426
44444427
44444428
44444429
44444430
44444431
44444432
44444433
44444434
44444435
44444436
44444437
44444438
44444439
44444440
44444441
44444442
44444443
44444444
44444445
44444446
44444447
44444448
44444449
444444410
444444411
444444412
444444413
444444414
444444415
444444416
444444417
444444418
444444419
444444420
444444421
444444422
444444423
444444424
444444425
444444426
444444427
444444428
444444429
444444430
444444431
444444432
444444433
444444434
444444435
444444436
444444437
444444438
444444439
444444440
444444441
444444442
444444443
444444444
444444445
444444446
444444447
444444448
444444449
4444444410
4444444411
4444444412
4444444413
4444444414
4444444415
4444444416
4444444417
4444444418
4444444419
4444444420
4444444421
4444444422
4444444423
4444444424
4444444425
4444444426
4444444427
4444444428
4444444429
4444444430
4444444431
4444444432
4444444433
4444444434
4444444435
4444444436
4444444437
4444444438
4444444439
4444444440
4444444441
4444444442
4444444443
4444444444
4444444445
4444444446
4444444447
4444444448
4444444449
44444444410
44444444411
44444444412
44444444413
44444444414
44444444415
44444444416
44444444417
44444444418
44444444419
44444444420
44444444421
44444444422
44444444423
44444444424
44444444425
44444444426
44444444427
44444444428
44444444429
44444444430
44444444431
44444444432
44444444433
44444444434
44444444435
44444444436
44444444437
44444444438
44444444439
44444444440
44444444441
44444444442
44444444443
44444444444
44444444445
44444444446
44444444447
44444444448
44444444449
444444444410
444444444411
444444444412
444444444413
444444444414
444444444415
444444444416
444444444417
444444444418
444444444419
444444444420
444444444421
444444444422
444444444423
444444444424
444444444425
444444444426
444444444427
444444444428
444444444429
444444444430
444444444431
444444444432
444444444433
444444444434
444444444435
444444444436
444444444437
444444444438
444444444439
444444444440
444444444441
444444444442
444444444443
444444444444
444444444445
444444444446
444444444447
444444444448
444444444449
4444444444410
4444444444411
4444444444412
4444444444413
4444444444414
4444444444415
4444444444416
4444444444417
4444444444418
4444444444419
4444444444420
4444444444421
4444444444422
4444444444423
4444444444424
4444444444425
4444444444426
4444444444427
4444444444428
4444444444429
4444444444430
4444444444431
4444444444432
4444444444433
4444444444434
4444444444435
4444444444436
4444444444437
4444444444438
4444444444439
4444444444440
4444444444441
4444444444442
4444444444443
4444444444444
4444444444445
4444444444446
4444444444447
4444444444448
4444444444449
44444444444410
44444444444411
44444444444412
44444444444413
44444444444414
44444444444415
44444444444416
44444444444417
44444444444418
44444444444419
44444444444420
44444444444421
44444444444422
44444444444423
44444444444424
44444444444425
44444444444426
44444444444427
44444444444428
44444444444429
44444444444430
44444444444431
44444444444432
44444444444433
44444444444434
44444444444435
44444444444436
44444444444437
44444444444438
44444444444439
44444444444440
44444444444441
44444444444442
44444444444443
44444444444444
44444444444445
44444444444446
44444444444447
44444444444448
44444444444449
444444444444410
444444444444411
444444444444412
444444444444413
444444444444414
444444444444415
444444444444416
444444444444417
444444444444418
444444444444419
444444444444420
444444444444421
444444444444422
444444444444423
444444444444424
444444444444425
444444444444426
444444444444427
444444444444428
444444444444429
444444444444430
444444444444431
444444444444432
444444444444433
444444444444434
444444444444435
444444444444436
444444444444437
444444444444438
444444444444439
444444444444440
444444444444441
444444444444442
444444444444443
444444444444444
444444444444445
444444444444446
444444444444447
444444444444448
444444444444449
4444444444444410
4444444444444411
4444444444444412
4444444444444413
4444444444444414
4444444444444415
4444444444444416
4444444444444417
4444444444444418
4444444444444419
4444444444444420
4444444444444421
4444444444444422
4444444444444423
4444444444444424
4444444444444425
4444444444444426
4444444444444427
4444444444444428
4444444444444429
4444444444444430
4444444444444431
4444444444444432
4444444444444433
4444444444444434
4444444444444435
4444444444444436
4444444444444437
4444444444444438
4444444444444439
4444444444444440
4444444444444441
4444444444444442
4444444444444443
4444444444444444
4444444444444445
4444444444444446
4444444444444447
4444444444444448
4444444444444449
44444444444444410
44444444444444411
44444444444444412
44444444444444413
44444444444444414
44444444444444415
44444444444444416
44444444444444417
44444444444444418
44444444444444419
44444444444444420
44444444444444421
44444444444444422
44444444444444423
44444444444444424
44444444444444425
44444444444444426
44444444444444427
44444444444444428
44444444444444429
44444444444444430
44444444444444431
44444444444444432
44444444444444433
44444444444444434
44444444444444435
44444444444444436
44444444444444437
44444444444444438
44444444444444439
44444444444444440
44444444444444441
44444444444444442
44444444444444443
44444444444444444
44444444444444445
44444444444444446
44444444444444447
44444444444444448
44444444444444449
444444444444444410
444444444444444411
444444444444444412
444444444444444413
444444444444444414
444444444444444415
444444444444444416
444444444444444417
444444444444444418
444444444444444419
444444444444444420
444444444444444421
444444444444444422
444444444444444423
444444444444444424
444444444444444425
444444444444444426
444444444444444427
444444444444444428
444444444444444429
444444444444444430
444444444444444431
444444444444444432
444444444444444433
444444444444444434
444444444444444435
444444444444444436
444444444444444437
444444444444444438
444444444444444439
444444444444444440
444444444444444441
444444444444444442
444444444444444443
444444444444444444
444444444444444445
444444444444444446
444444444444444447
444444444444444448
444444444444444449
4444444444444444410
4444444444444444411
4444444444444444412
4444444444444444413
4444444444444444414
4444444444444444415
4444444444444444416
4444444444444444417
4444444444444444418
4444444444444444419
4444444444444444420
4444444444444444421
4444444444444444422
4444444444444444423
4444444444444444424
4444444444444444425
4444444444444444426
4444444444444444427
4444444444444444428
4444444444444444429
4444444444444444430
4444444444444444431
4444444444444444432
4444444444444444433
4444444444444444434
4444444444444444435
4444444444444444436
4444444444444444437
4444444444444444438
4444444444444444439
4444444444444444440
4444444444444444441
4444444444444444442
4444444444444444443
4444444444444444444
4444444444444444445
4444444444444444446
4444444444444444447
4444444444444444448
4444444444444444449
44444444444444444410
44444444444444444411
44444444444444444412
44444444444444444413
44444444444444444414
44444444444444444415
44444444444444444416
44444444444444444417
44444444444444444418
44444444444444444419
44444444444444444420
44444444444444444421
44444444444444444422
44444444444444444423
44444444444444444424
44444444444444444425
44444444444444444426
44444444444444444427
44444444444444444428
44444444444444444429
44444444444444444430
44444444444444444431
44444444444444444432
44444444444444444433
44444444444444444434
44444444444444444435
44444444444444444436
44444444444444444437
44444444444444444438
44444444444444444439
44444444444444444440
44444444444444444441
44444444444444444442
44444444444444444443
44444444444444444444
44444444444444444445
44444444444444444446
44444444444444444447
44444444444444444448
44444444444444444449
444444444444444444410
444444444444444444411
444444444444444444412
444444444444444444413
444444444444

5

and may be configured with selected thicknesses to provide a plurality of desired reaction zones or gaps therebetween. With a plurality of gaps, the total non-thermal plasma environment is expanded to increase the overall flow rate of the exhaust gas or throughput of the apparatus.

In another embodiment, a plurality of double-dielectric electrodes may be supported on a specially configured fluoropolymeric (e.g., fluorocarbon) insulators adapted for placement inside the plasma reactor. In particular, the insulators may be configured with grooves or indentations which support the double-dielectric electrodes at their edge portions, in parallel formation, alternating between positive and negative charges. The grooves may be configured to be spaced apart at selected distances such that adjacent double-dielectric electrodes may provide a plurality of desired reaction zones or gaps. Again, with a plurality of gaps, the total non-thermal plasma environment is expanded to increase the overall flow rate of the exhaust gas or throughput of the apparatus.

20

25

The non-thermal plasma apparatus of the instant invention may provide for the pretreatment of the exhaust gas with ethanol, either by vapor absorption or direct vapor injection. In particular, before the exhaust gas is exposed to plasma reactor, at least a portion of the exhaust gas is

5

exposed to ethanol before entering the plasma reactor. One method involves diverting at least a portion of the exhaust gas through an ethanol bath before the exhaust gas enters the plasma reactor. Another method involves directly injecting ethanol directly into the path of the exhaust gas before it enters the plasma reactor.

For either method, the high vapor pressure of ethanol permits a significant portion of the ethanol to be absorbed by the exhaust gas. As such, the ethanol-bearing exhaust gas is pretreated in preparation for the plasma reactor. In particular, the plasma reactor exposes to the pretreated exhaust gas to reactive species generated by the plasma reactor, e.g., oxygen atoms, whereupon nitric oxides are converted to a variety of products, including primarily nitrogen dioxide, with significantly improved efficiency. As an added advantage, the solubility of ethanol permits the ethanol to be readily scrubbed from the exhaust gas downstream of the plasma reactor, along with the converted nitrogen dioxide.

20

To implement the ethanol pretreatment, the non-thermal plasma apparatus comprises an inlet and an outlet connected to the reaction chamber, permitting the exhaust gas to enter and leave the reaction chamber. In one embodiment, the inlet is further connected to a diverter equipped with an

40
39
38
37
36
35
34
33
32
31
30
29
28
27
26
25
24
23
22
21
20
19
18
17
16
15
14
13
12
11
10
9
8
7
6
5
4
3
2
1

injector, which diverts a portion of the gas stream through an ethanol bath before reinjecting the ethanol-bearing gas stream into the inlet. Ethanol is readily absorbed by the gas stream as it passes through the ethanol bath.

5

In another embodiment, the inlet is equipped with an injector which receives a supply of ethanol that is sprayed as a fine mist directly into the gas stream. The fine mist of ethanol is substantially uniformly absorbed by the gas stream before it enters the reactor chamber. A reservoir stores the ethanol which is delivered to the injector by a metered pump.

The non-thermal plasma treatment may be used for a variety of gas streams. A typical gas stream contains approximately nitrogen, oxygen, water vapor and nitric oxide. The primary function of the treatment is to convert the nitric oxide into nitrogen dioxide.

20

These, as well as other features of the invention, will become apparent from the detailed description which follows, considered together with the appended drawings.

DESCRIPTIONS OF THE DRAWINGS

In the drawings, which constitute a part of this specification, exemplary embodiments demonstrating various features of the invention are set forth as follows:

5 FIGURE 1 is a perspective view of an embodiment of a non-thermal plasma apparatus;

FIGURE 1B is a partial perspective view of an embodiment of a dielectrically-coated (double-dielectric) electrode;

FIGURE 2 is a perspective view of another embodiment of a non-thermal plasma apparatus;

FIGURES 3A and 3B are a side elevational view and an end elevational view of an embodiment of a non-thermal plasma reactor utilizing double-dielectric electrodes;

20 FIGURES 4A and 4B are a side, partially exploded, elevational view and an end elevational view of another embodiment of a non-thermal plasma reactor utilizing double-dielectric electrodes; and

25 FIGURE 5 is a graph of NO concentration as function of power of a non-thermal plasma apparatus.

DESCRIPTION OF THE ILLUSTRATIVE EMBODIMENTS

As indicated above, detailed illustrative embodiments are disclosed herein. However, structures for accomplishing the objectives of the present invention may be detailed quite differently from the disclosed embodiments. Consequently, specific structural and functional details disclosed herein are merely representative; yet, in that regard, they are deemed to afford the best embodiment for purposes of disclosure and to provide a basis for the claims herein which define the scope of the present invention.

Referring to FIGURE 1A, a non-thermal plasma apparatus 10 for treating effluent or exhaust gas 11 containing nitric oxides ("NO_x") is illustrated. The apparatus includes a plasma reactor 12 having a plasma chamber or reaction chamber 14, connected to a voltage supply 16. An inlet 18 (shown partially broken away) and an outlet 20 are connected to opposing ends of the chamber 14, allowing the gas 11 to enter and exit the chamber 14. A scrubber 22 is provided below or downstream from the plasma reactor 12, for the removal of soluble constituents from the gas 11. The scrubber 22 may ultimately lead to a stack 24 to release the exhaust gas 11 from the apparatus 10.

20
21
22
23
24
25
26
27
28
29
30

treatment may contain 82% nitrogen, 15% oxygen, 3% water vapor, and approximately 1000 ppm nitric oxide (NO). A primary function of the apparatus 10 is to convert the nitric oxide (NO) into nitrogen dioxide (NO₂). In certain instances, particularly where a preinjectant such as ethanol is used, the nitric oxide (NO) may be converted to primarily to nitrogen dioxide (NO₂) and some nitric acid (HNO₃). In addition, small amounts of the compounds peroxy acetyl nitrate, acetaldehyde, and n-propyl nitrate may also be produced.

20
25
30
35
40
45
50
55
60
65
70
75
80
85
90
95

The desired conversions are driven by electro-chemical reactions facilitated by the plasma reactor 12 of the apparatus 10. These reactions are non-thermal or "cool" reactions in that the dispersed electron charge or electric field (i.e., a "corona") generated by the plasma reactor energizes primarily the electrons, substantially leaving the constituents of the gas relatively unenergized or "unheated." As such, the energized electrons enhance the electron density of the gas as it travels through the plasma reactor 12, promoting electron-molecule collision, as opposed to molecule-molecule collision. Such charge dispersion and electron concentration drive the desired conversion of nitric oxides to other atomic and molecular forms, either chemically less toxic, and/or structurally better suited for removable or separation from the gas stream 11.

In accordance with a feature of the present invention, the plasma reactor 12 includes at least a pair of dielectrically-coated electrodes 26a and 26b and defining a non-thermal plasma environment or reaction zone 28 therebetween. As shown in FIGURE 1B, the electrode has a planar configuration and includes a metal electrode plate 30 covered with a coating 32 of a fluoropolymeric substance, for example, fluorocarbon. The coating 32 isolates the electrode plate 30 from the exhaust gas 11 and its nonconductive nature enables it to standoff a defined level of voltage that is applied to the electrode plate. The voltage is applied by the voltage 16 supply via a wire 34 that is attached to the plate 30 and extends through the coating 32. In contrast to conventional materials from which dielectrics may be made (e.g., such as quartz, glass, alumina, mullite, and oxide free ceramic such as silicon nitrite, boron nitrite, aluminum nitrite), fluoropolymers, and especially fluorocarbons, may be readily applied to the surface of the electrode plate 30 to constitute the coating 32. The thickness of the coating 32 may vary depending on the level of contamination of the gas and the flow rate. Fluorocarbon tapes, commercially available under the names of Teflon®, Teflon® PFA, and Dykor® may be applied directly to the electrode plates. Moreover, Teflon® PFA - coated plate electrodes are available from Toefco Engineering, Inc., Niles, Michigan.

0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150

20

25

The apparatus 10 utilizing the coated electrodes 26a and 26b has improved overall durability, when compared to conventional non-thermal plasma devices. The relatively high tensile strength of fluoropolymers, in particular, fluorocarbon, renders the coated electrodes more ductile and therefore offers greater protection against breakage.

Significantly, the apparatus 10 offers improved durability with relatively little change in electrical and thermal performance.

The plasma reactor 12 may utilize more than the coated electrodes 26a and 26b. In particular, as illustrated in FIGURE 1, the reaction chamber 14 may house a plurality of coated electrodes 26i which are arranged in parallel formation, alternating between positively-charged and negatively-charged electrodes. The number of electrodes also varies depending on the level of contamination in the gas and the flow rate of the gas.

It can be seen that the electrodes 26i are each coated on both surfaces of the underlying plate 30i. Accordingly, the plates of adjacent alternating high and low electrodes remain coated and isolated from the gas. These electrodes may be referred to as "double-dielectric" electrodes. The voltage supply 16 may supply voltage to all the electrodes, such that the reaction zone 28 of the apparatus is expanded to include all gaps 36 in between each adjacent

20
15
10
5
P07 39512

20
25

pairs of high and low electrodes 26i. Such a voltage supply is available from Elgar Corporation, San Diego, California, under part #1001SL-11.

5 In operation, the exhaust gas 11 may be forced or drawn through the apparatus 10 by an induction fan (now shown), including through the inlet 18, the plasma chamber 14, the outlet 18, the scrubber 22 and the stack 24, as understood by those of ordinary skill in the art. The exhaust gas 11 enters the apparatus 10 through the inlet 18 which terminates at the plasma reactor 12. Once inside the plasma chamber 14 of the plasma reactor 12, the gas 11 is exposed to the electric field between the electrodes 26i of the reactor 12 as generated by the selected voltage applied by the voltage supply 16. The strength of the electric field is above the critical field strength of the exhaust gas 11, but not so high as to establish a condition conducive to sustain arcing between the electrodes 26i. In particular, the selected voltage creates a multitude of short-lived current filaments or breakdown channels 20 (commonly called microdischarges) within the gas inside the reaction zone 28.

25 Regardless of the configuration of the electrodes, the flow of electrons at high velocity and in high density increase the likelihood of an NO_x reduction producing collision. Specifically, after a discharge extinguishes, the

reactive species, e.g., oxygen atoms, diffuse out of the channels into the rest of the gas stream in the reaction zone 28 (i.e., in between each of the gaps 36) where they drive the desired conversions. Because the energy of the discharge is used to directly accelerate the electrons, rather than heating the gas 11, the apparatus 10 provides an attractive technique for selectively driving specific chemical reactions. In this regard, the apparatus 10 has applications beyond the specific chemical reaction discussed herein.

Referring to FIGURES 3A and 3B, a planar, variable gap, non-thermal plasma reactor 40 for use with the apparatus 10 is illustrated. In particular, two to five double-dielectric electrodes 42i are provided in the reactor's chamber 44 and arranged in parallel formation such that the gas 11 is directed through gaps 46 between the electrodes 42i.

The chamber 44 in FIGURES 3A and 3B is constructed of aluminum with penetrations for the inlet 48 and the outlet 50 and high and low voltage terminals 52. Such inlet/outlet coupling and connection devices are commercially available from The Swagelok Companies, Solon, Ohio. The interior of the chamber is overlaid with Kapton® high voltage tape and the upper portion is sealed with a rubber gasket 54 and a Plexiglas cover 56. The electrodes 42i may range between approximately 6" to 8" wide by approximately 22" to 24" long, by

approximately 0.09" to 1.25" thick. The electrodes 42i are configured to alternate between positive (high) or negative (low) charges and are separated by fluorocarbon spacers 58 to provide the desired gaps 46. The thickness of the 5 fluoropolymer-based coating 32 on the electrodes, as well as the thickness of the spacers 58, may be varied to vary the width of the gaps 46. A suitable gap width may range between approximately 0.5 to 3.0 mm, rendering the total active volume of the reaction zone of the plasma reactor 40 to range between 50.0 to 1,500 ml.

Depending on the capacity of the reactor 40, the flow rate of the apparatus may vary up to approximately 400 cubic feet per minute. As such, the residence time of the mixed gas in the reaction chamber 44 may range between 0.1 to 60.0 seconds, for example, approximately 0.3 seconds.

Another plasma reactor 64 that may be used with the apparatus 10 is illustrated in FIGURES 4A and 4B. A reactor 20 chamber 66 of the reactor 64 is again constructed of aluminum and defines an interior region measuring approximately 8" by 8". The chamber 66 is approximately 31" in length and is fitted with an inlet 68 and an outlet 70. Silicone rubber gaskets 72 and end caps 74 (one shown in exploded view) are provided. A glass window (not shown) may be fitted in the end 25 caps included to permit viewing of the interior of the chamber.

Ceramic and copper high voltage terminals 76 are also provided.

Insulating the interior of the reactor chamber 66 is a fluorocarbon member 78 configured with grooves 82 of selected dimensions to receive and support edge portions of double-dielectric electrodes 80 in a desired arrangement. Different insulating members may be used with the reactor 64, configured with different separation distances between the grooves 82. Accordingly, different desired gap dimensions may be attained. Supported on its edge portions, the electrodes 80 can readily slide in and out of the chamber 66. The electrodes 80 are arranged in alternating high and low configuration and connected to the high voltage terminals 76 at opposite ends of the chamber 66.

It bears emphasis that the plasma reactor of the apparatus 10 is non-thermal in that there is a cool discharge promoting the free flow of electrons. Significantly, only the electrons gain appreciable energy in the reaction zone(s) or gap(s) and hence increased temperature is experienced primarily by the electrons, leaving the remaining constituents of the gas stream substantially unheated. The energy is generally evenly distributed over the non-thermal plasma environment; any hot discharge is typically a localized point to point arc.

Table 1 shows selected properties of one type of fluorocarbon (i.e., PFA Teflon®) compared to Pyrex® glass.

| Material | Dielectric Constant | Dielectric Strength (kV/mm) | Tensile Strength (kpsi) | Working Temp. (C) |
|--------------|---------------------|-----------------------------|-------------------------|-------------------|
| PFA Teflon® | 2.1 | 60 | 2.20 | 260 |
| Pyrex® glass | 4.5 | 13 | 0.14 | 450 |

While the dielectric constant of PFA Teflon® is comparable to that of Pyrex® glass, the dielectric strength is much higher. In view of this observation, the thickness of a PFA Teflon® coating can be less than that used for Pyrex®, leaving more volume for the gas stream. The desirable working temperature of PFA Teflon® is lower than that for Pyrex®, however, it remains sufficient for treating most types of contaminated gas. As previously noted, the PFA Teflon® provides relatively high tensile strength, rendering the electrodes ductile and less susceptible to breakage, thus improving the efficiency and durability of the apparatus.

Experimental results presented in Table 2. also demonstrates an improvement in efficiency when the apparatus 10 is used to treated NO-bearing exhaust stream from a diesel generator.

Table 2. Increased Efficiency of Fluorocarbon Dielectric in NO Conversion

| Material | Power (Watts) | Flow (liters per minute) | Percentage Conversion NO to NO ₂ | eV/molecule Converted |
|--------------|---------------|--------------------------|---|-----------------------|
| PFA Teflon® | 48 | 100 | 100 | 17 |
| Pyrex® Glass | 48 | 100 | 66 | 22 |

10

As also mentioned, the addition of ethanol (C₂H₅OH) into the exhaust gas significantly enhances the efficiency of the conversion in terms of the electrical power used to drive the conversion.

Referring back to FIGURE 1, an ethanol bath 84 is provided to improve the efficiency of the conversion process. A diverter 86 is connected to the inlet 18, having a first section 86a extending from the inlet 18 to the ethanol bath 84 and a second section 86b extending from the ethanol bath 84 to the inlet 18. At the junction between the second section 86a and the inlet 18, an injector 88 is provided above or upstream from the plasma reactor 12. In operation, the apparatus 10 diverts a portion of the exhaust gas 11 through the diverter 86 to pass through the ethanol bath 84. Due to the high vapor pressure of ethanol (C₂H₅OH), the ethanol is vaporized along with and mixed into the gas stream. As the ethanol-bearing gas

SEARCHED
INDEXED
SERIALIZED
FILED

25

reaches the injector 88, it is injected into the inlet 18 to mix with the gas in the inlet.

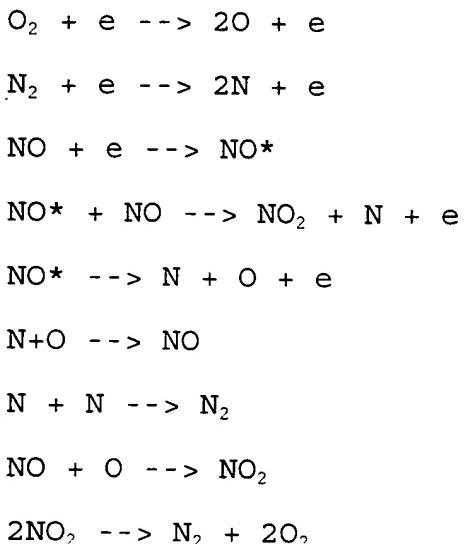
Referring to FIGURE 2, an alternative embodiment of
5 the non-thermal plasma apparatus 10 is illustrated. The injector 88 remains upstream from the plasma reactor 12, but is fed directly with ethanol stored in a reservoir 90 and delivered via a metered pump 92. The ethanol is injected into the gas in the inlet as a fine mist that evaporates substantially uniformly into the gas stream 11. A small varistaltic pump available from Manostat Corporation, Barrington, Illinois, under Model #72-305-000, may be used.

Referring to both FIGURES 1 and 2, as the gas stream (ethanol-bearing or not) approaches the plasma reactor 12, a non-thermal plasma environment is prepared in the reaction chamber 14. The non-thermal plasma environment of the reaction zone 28 contains generated reactive agents, in particular, energetic electrons, to drive selected electrical and/or chemical reactions in the gas, which are discussed in detail further below.

It has been observed that low energy gaseous plasma exhibit physical and chemical properties different than their normal properties. One such property is extreme chemical reactivity, which as discussed above, involves free electrons

being energized in the electrical field or corona until the electrons attain sufficient energy to cause ionization of some of the gas molecules. Reactions for NO_x reduction using the free electron e may be as follows:

5



With the preinjection of ethanol (C₂H₅OH) in accordance with feature of the present invention, several additional molecular compounds are possible, such as nitric acid (HNO₃) and even small amounts of peroxy acetyl nitrate, acetaldehyde, and n-propyl nitrate.

20

The use of ethanol as a pre-injectant increases the efficiency of the non-thermal plasma apparatus by as much as a factor of ten. The combined use of ethanol injection and fluorocarbon dielectrics further increases efficiency.

25

Referring to FIGURE 5, the NO concentrations in gas streams

containing molar ratios of ethanol to initial NO of 8:1 are compared for Pyrex® glass and Teflon® PFA dielectrics. When using the Teflon® PFA dielectrics, substantially 90% NO removal was obtained with only 48 Watts of plasma reactor power. The calculated molecular energy consumption for this data point was approximately 17 eV per NO representing a 30% decrease in energy consumption as compared to the stream treated with Pyrex® glass dielectrics.

Improvements in efficiencies were observed when treating NO_x contained in diesel generator exhaust. Referring to Table 1, results of an untreated stream were compared with results of a stream treated with approximately 4.0 ml/min. of ethanol injectant.

Table 3. Ethanol Injection Performance in Diesel Exhaust Stream

| Condition | Percent NO Reduction | Electron Volts/Molecule |
|-----------------|----------------------|-------------------------|
| Ethanol Treated | 99.6 | 17.3 |
| Untreated | 17.3 | 170.4 |

As also mentioned, the ethanol offering a high vapor pressure and solubility in water offers ease in application. Its characteristic high vapor pressure renders it to be readily injected into the gas stream in either vapor or liquid form. Its high solubility in water also ensures that it may be

readily scrubbed from the system along with the converted nitrogen dioxide (NO_2) from the exiting gas stream.

In comparison to other preinjection compounds, an increase in efficiency may be achieved, however, not without increased complication in the overall process and apparatus. Many hydrocarbons have a lower vapor pressure which makes injection into the gas stream difficult except in higher temperature conditions. Also, as the gas stream cools, the injectant tends to condense on the surface of the electrodes shorting out the voltage circuit. Other possible injectants are not water soluble and therefore complicates the scrubbing process.

As illustrated in FIGURES 1 and 2, the apparatus is pack-free, that is, free of any additional dielectric material, such as glass wool packing. Accordingly, the gas stream flows through the reaction zone(s) or gap(s) substantially unimpeded.

It may be seen that the system of the present invention may be readily incorporated in various embodiments to provide a non-thermal plasma treatment of effluent or exhaust gas. It is understood by one of ordinary skill in the art that the voltage applied, the frequency of the voltage applied, the gas stream flow rate, temperature and residence time in the reactor chamber may affect power consumption and therefore

efficiency of the apparatus. It is also understood by one of ordinary skill in the art that the configurations and dimensions of various components, such as the plasma chamber, the electrodes, the spacers and the insulation may affect the efficiency and throughput of the apparatus. For example, the electrodes may be variously configured as two dimensional or three dimensional conductive elements. They may be rectilinear or tubular, or combinations thereof. Any one or combinations of these parameters may be tailored to generate specific reactive species and/or to target specific constituents in the exhaust gas for conversion. In that regard, various alternative techniques and configures may be employed departing from those disclosed and suggested herein.

Consequently, it is to be understood that the scope hereof should be determined in accordance with the claims as set forth below.